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(54) ELECTROCONDUCTIVE RESIN COMPOSITION AND MOLDED ARTICLE

(57)Abstract:

PROBLEM TO BE SOLVED: To obtain a thermoplastic resin molded article containing an electroconductive material, which is suppressed in deterioration of impact strength caused by incorporation of an electroconductive material component and has excellent surface appearances (excellent gloss and no flow marks) and a satisfactory surface resistivity.

SOLUTION: The electroconductive resin composition comprises 10-80 wt.% of a polypropylenic resin obtained by homopolymerization of propylene or random copolymerization of propylene with ethylene, 5-50 wt.% of a linear low density polyethylene as a modifier component obtained by polymerization using a metallocene catalyst, and 5-70 wt.% of at least one electroconductive material selected among carbon black, graphite, metal powder and whisker thereof, metal fiber, metal oxide and carbon fiber. The molded article is obtained using the same.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the impact strength excellent in polypropylene resin mold goods, and the conductive resin constituent which gives high glossiness.

[0002]

[Description of the Prior Art] Making thermoplastics, especially a polypropylene resin contain electrical conducting materials including carbon black, and lecturing on electrostatic preventive measures is applied to various kinds of thermoplastics mold goods centering on the electronic parts which are easy to receive band electrical-potential-difference destruction. It has many descriptions -- that specific gravity of a polypropylene resin is small, that chemical resistance is good, cheap being a thing, and a moldability is good -- and various mold goods including the electric conduction container using a polypropylene resin, a sheet, a tray, and a pipe are supplied widely.

[0003]

[Problem(s) to be Solved by the Invention] However, since 10 - 25% of the weight of addition is needed in a mold-goods constituent if it is a remarkable addition, for example, carbon black, in order to make conductivity discover when electrical conducting materials are carbon black, a metal powder, and conductive metal oxidization powder, there is a fault to which the fluidity which the polypropylene resin originally has, and shock resistance fall sharply.

[0004] As technique used from the former in order to conquer this fault, in a resin constituent, ethylene-propylene rubber (it omits Following EPR), Or although adding the polyethylene of olefin system rubber, such as ethylene propylene diene rubber (it omits Following EPDM), or the amount of giant molecules, and raising shock resistance is performed widely A fluidity and the surface gloss of mold goods got worse, and when it was especially an injection-molded product, there were a short shot (poor shaping phenomenon produced in order that resin may not flow to the back of a molding die), the flow mark, and a fault that the appearance of mold goods got worse by surface cloudiness.

[0005]

[Means for Solving the Problem] this invention person in the impact amelioration agent by conventional olefin system rubber, polyethylene, etc. Without reducing the conductivity of the polypropylene resin containing an electrical conducting material, and shock resistance A fluidity, An example is taken by the above-mentioned fact of holding the gloss on the front face of mold goods and that the technical problem which the conductive resin constituent of an antinomy holds is unsolvable. The result of having examined wholeheartedly the new impact amelioration agent to which amelioration of a resin fluidity, an improvement of compatibility with a polypropylene resin, and the glossiness on the front face of mold goods may be given, By adding in a conductive resin constituent by making into an impact amelioration agent the straight chain-like low density polyethylene (it abbreviating to Metallocene LLDPE below) by which the polymerization was carried out using the metallocene catalyst, it resulted in amelioration of the resin fluidity which was not acquired and surface gloss with the conventional configuration. When the property over the system using the EPDM content polypropylene resin of olefin

system rubber used especially conventionally was verified, in the combination system which has the same Izod impact strength, it was checked that the fluidity and surface gloss improve by leaps and bounds.

[0006] That is, invention of the 1st of this invention is 10 - 80 % of the weight of polypropylene resins obtained by propylene homopolymerization or the random copolymerization of a propylene and ethylene, 5 - 50 % of the weight of straight chain-like low density polyethylene by which the polymerization was carried out, using a metallocene catalyst as a modifier component, carbon black, graphite, a metal powder and its whisker, a metal fiber, a metallic oxide, and a conductive resin constituent characterized by including 5 - 70 % of the weight of at least one or more kinds of electrical conducting materials chosen from a carbon fiber.

[0007] The 2nd invention is a conductive resin constituent given in the 1st invention characterized by an electrical conducting material being carbon black.

[0008] It is a conductive resin constituent given in the 1st or 2nd invention characterized by the 3rd invention having straight chain-like low density polyethylene by which the polymerization was carried out using the metallocene catalyst in the range of the melt flow rate measured value of 1-100g / 10min. in 230 degrees C and 2160g load.

[0009] the 1-3rd invention characterized by the 4th invention having straight chain-like low density polyethylene by which the polymerization was carried out using the metallocene catalyst in the range of a consistency 840 - 920 kg/m³, and bending elastic-modulus 100-500MPa -- it is the conductive resin constituent of a publication either.

[0010] the 5th invention -- the 1-4th invention -- they are the mold goods obtained using the conductive resin constituent of a publication either.

[0011]

[Embodiment of the Invention] Hereafter, this invention is explained concretely. As a polypropylene resin used for this invention, it is JIS. The resin which can measure the melt flow rate based on K7210 and which is generally used corresponds. For example, although the resin by propylene homopolymerization, the resin by the random copolymerization of a propylene and ethylene, etc. are mentioned, since the wax-like polymer with melt flow rate measurement difficult since the surface gloss of mold goods gets worse and the thing which made a block copolymer and olefin system rubber with ethylene contain becomes poor [an appearance] for shock-proof improvement cannot discover the mechanical strength as mold goods, it is removed.

[0012] Moreover, the melt flow rate of the polypropylene resin concerned (it measures by the approach specified to JIS K7210.) As measured value of the measurement temperature of 230 degrees C, and 2160g load of measuring loads, 0.1-10g/10min., and the object for injection-molded products have [a pipe, a sheet, and the object for film mold goods] the desirable range of 1-100g / 10min.

[0013] As an electrical conducting material used for this invention, carbon black, graphite, a metal powder and its whisker, a metal fiber, a metallic oxide, a carbon fiber, etc. are mentioned, and these are independent or can be used in two or more sorts of combination. Conductivity and in order to make especially an antistatic function provide and to get, in a surface-electrical-resistance value, the engine performance of 1 M ohm or less is needed. therefore -- although powdered materials, such as carbon black, graphite, and a metal powder, change with specific gravity of an electrical conducting material -- the case of carbon black -- 8 - 30 % of the weight -- in the case of graphite or a metal powder, 40 - 60 % of the weight needs to be added preferably 30 to 75% of the weight ten to 25% of the weight. In the case of a whisker and a fibrous object, it is possible to make it decrease from the above-mentioned addition. Moreover, in the case of stainless steel fiber and a carbon fiber, it is possible to set a surface-electrical-resistance value to 1 M ohm or less at 5 - 15 % of the weight.

[0014] As an impact amelioration agent used for this invention, the polyethylene by which the polymerization was carried out with the metallocene catalyst is used. This resin is the copolymer of polyethylene and alpha-olefin and 1-butene, 1-hexene, and 1-octene are mentioned as an alpha-olefin. Although the polymerization of the polyethylene used as the above-mentioned impact amelioration agent is cheaply carried out with a metallocene catalyst in recent years and expansion to various

applications is achieved Since especially the metallocene LLDPE of low degree of crystallinity can control molecular weight distribution narrowly and low-molecular-weight objects can be reduced efficiently, The fall beyond the stickiness nature on account of low degree of crystallinity and the need for the melting point, the emitting smoke prevention at the time of shaping, etc. have many descriptions, such as high gloss and a good fluid manifestation, as compared with other thermoplastic elastomer, and are used as the modifier of a polypropylene resin, and an impact amelioration agent in recent years.

[0015] The reason which the shock resistance which was excellent by using Metallocene LLDPE, surface glossiness, and a fluidity discover is considered as follows. The reason for having the shock resistance Metallocene LLDPE excelled [shock resistance] in the 1st is for during the detailed crystal-crystal being combined by existence of the Thailand molecule in ordinary temperature, becoming the false three-dimension network structure, and taking the same structure as other rubberlike substance, although Metallocene LLDPE is resin non-constructing a bridge. Although straight chain-like low density polyethylene usual in existence of the Thailand molecule is also checked, it is thought that Metallocene LLDPE has many above-mentioned tie molecule's existence probabilities as two to 4 times, and shock resistance tends [so] to discover it.

[0016] Although it is a reason for having the gloss which was excellent while Metallocene LLDPE had shock resistance in the 2nd, since there is no matter which has the dispersed particle diameter of a number which is seen with a polypropylene block copolymer - 10 microns of numbers, in the case of Metallocene LLDPE, with other olefin system rubber, it thinks because high gloss can be given to rubber dispersed particle diameters being a number - 10 microns of numbers, without forming irregularity in a mold-goods front face.

[0017] Although it is a reason for having the fluidity Metallocene LLDPE excelled [fluidity] in the 3rd, the Thailand molecule which forms the three-dimension network structure stated by the 1st term is because it is possible to take the same behavior as usual polyethylene near the molding temperature, in order to disappear with fusion of a crystal by being heated, although it exists in ordinary temperature. On the other hand, by the elastomer containing olefin system rubber, even if it is near the molding temperature, it is thought that it has the inclination for a fluidity to also get worse as rubber loadings increase since rubber remains as a bad fluid particle which constructed the bridge.

[0018] The metallocene LLDPE used by this invention is resin by which a polymerization is carried out with a metallocene catalyst, and a metallocene catalyst is the name of the catalyst which at least one or more ligands which have a cyclopentadienyl frame in tetravalent transition metals, such as titanium, a zirconium, nickel, palladium, a hafnium, niobium, and platinum, configure concretely.

[0019] As ligand which has a cyclopentadienyl frame A cyclopentadienyl group, a methylcyclopentadienyl radical, an ethylcyclopentadienyl radical, n- or i-propylcyclopentadienyl radical, n-, i-, sec-, tert-, a butylcyclopentadienyl radical, a hexyl cyclopentadienyl group, Alkyl 1 substituent cyclopentadienyl groups, such as an octyl cyclopentadienyl group, A dimethylcyclopentadienyl radical, a methylethyl cyclopentadienyl group, A methylpropyl cyclopentadienyl group, a methylbutyl cyclopentadienyl group, A methylhexyl cyclopentadienyl group, an ethyl butylcyclopentadienyl radical, Alkyl 2 substituent cyclopentadienyl groups, such as an ethylhexyl cyclopentadienyl group, A trimethylcyclopentadienyl radical, a tetramethylcyclopentadienyl radical, Alkyl many substituent cyclopentadienyl groups, such as a pentamethylcyclopentadienyl group, Cyclo permutation hexyl cyclopentadienyl groups, such as a methylcyclohexyl cyclopentadienyl group, an indenyl group, 4, 5 and 6, 7-tetrahydroindenyl radical, a fluorenyl group, etc. are mentioned.

[0020] As ligands other than the ligand which has a cyclopentadienyl frame, the anion ligand of monovalence, such as chlorine and a bromine, the anion chelate ligand of bivalence, a hydrocarbon group, an alkoxide, an amide, an aryl amide, aryl oxide, phosphide, aryl phosphide, a silyl radical, a permutation silyl radical, etc. are mentioned, for example. As the above-mentioned hydrocarbon group, a with a carbon number of about one to 12 thing is mentioned, for example, aralkyl radicals, such as aryl groups, such as cycloalkyl radicals, such as alkyl groups, such as a methyl group, an ethyl group, a propyl group, butyl, an isobutyl radical, an amyl group, an isoamyl radical, a hexyl group, a heptyl radical, an octyl radical, a nonyl radical, a decyl group the Cecil radical, a decyl group, the Cecil radical,

and a 2-ethylhexyl radical, a cyclohexyl radical, and a cyclopentyl group, a phenyl group, a tolyl group, benzyl, and a neo fill radical; a nonylphenyl radical, etc. be mentioned.

[0021] As a metallocene compound which the ligand which has a cyclopentadienyl frame configured Cyclopentadienyl titanium tris (dimethyl amide), bis(cyclopentadienyl) titanium dichloride, Dimethylsilyl tetramethylcyclopentadienyl-p-n-buthylphenyl amide zirconium chloride, Methylphenyl silyl tetramethylcyclopentadienyl-tert-butyl amide hafnium dichloride, Dimethylsilyl tetramethyl SHIKUROPENDA dienyl-tert-butyl amide hafnium dichloride, Indenyl SHITANIUMU tris (diethyl amide), an indenyl titanium bis(di-n-butyl amide) indenyl titanium screw (G n-propyl amide), etc. are mentioned.

[0022] These polymerizations can be performed by the catalyst system which added for example, methylamino KISAN, a boron compound, etc. as cocatalyst other than the metallocene system catalyst containing the above-mentioned tetravalent transition metals. In this case, as for the rate of these catalysts to a metallocene catalyst, it is desirable that it is one to 1 million time mol.

[0023] The above-mentioned conductive resin constituent can be manufactured by using the approach of making thermoplastics kneading various fillers. For example, it is attained by using continuous system kneading machines, such as batch type kneading machines, such as a pressurized kneader and a Banbury mixer, a monopodium and a biaxial extruder, a tandem-die kneading machine, and a ko kneader. Although it is possible to begin carbon black by these approaches and to knead the shape of various powder and a whisker and a fibrous electrical conducting material, it is important to perform kneading with resin, without destroying the configuration, in case the shape of a whisker and a fibrous electrical conducting material are kneaded.

[0024] In case it kneads, the antioxidant of resin, an ultraviolet ray absorbent, light stabilizer, lubricant, and a coloring agent may be added if needed. independent [because of the heat deterioration prevention at the time of processing of resin / in antioxidants, such as a phenol system, the Lynn system, a sulfur system, and a lactone system,] as an antioxidant -- or it can mix and add. Moreover, when weatherability is required, a benzophenone system, a SARUSHI rate system, a benzotriazol system, a cyanoacrylate system, a HINDATO amine system compound, etc. can be used as an ultraviolet ray absorbent or light stabilizer on an outdoor-type way. As lubricant used in order to perform slippage grant at the time of kneading and shaping, a polyolefine wax, a higher-fatty-acid metal salt or an amide, an ester compound, etc. can be added. What is necessary is just to use the thing of the powder which carried out dispersible processing of the pigment generally used for coloring of polypropylene as a coloring agent, a liquid, and a pellet type.

[0025]

[Example] This invention is explained in more detail using an example and the example of a comparison. Hereafter, the section indicates weight %s to be the weight section and %. Explanation of an ingredient, the presentation, and the property were indicated to the table.

After kneading 20 % of the weight of [example 1] carbon, 59.8 % of the weight of gays PP, 20 % of the weight of metallocenes LLDPE, and 0.2 % of the weight of anti-oxidants with a Banbury mixer, it corned on the cylindrical pellet with a diameter [of 3mm], and a die length of 3mm. After the obtained pellet measured the melt flow rate by 230 degrees C and 2160g load, it produced the 200mmx400mm plate with the injection molding machine of 75t of clamping pressure, and measured gloss and a surface-electrical-resistance value. Furthermore, the existence of the flow mark was checked visually. Moreover, the piece of an Izod test with a thickness of 5mm was fabricated to coincidence, and eye DODDO impact strength was measured.

[0026] The gay PP of the [example 2] example 1 was changed into Random PP, and also the same conduct as an example 1 was performed.

[0027] The gay PP of the [example 1 of comparison] example 1 was changed into Block PP, and also the same conduct as an example 1 was performed.

[0028] The pitch (Gay PP and Metallocene LLDPE) of the [example 2 of comparison] example 1 was changed into TPO, and also the same conduct as an example 1 was performed.

[0029] The metallocene LLDPE of the [example 3 of comparison] example 1 was changed into LLDPE

in which the polymerization was carried out by the Ziegler catalyst. Others performed the same conduct as an example 1.

[0030] After kneading 60 % of the weight of [example 3] metallic oxides, 29.8 % of the weight of gays PP, 10 % of the weight of metallocenes LLDPE, and 0.2 % of the weight of anti-oxidants with a Banbury mixer, it corned on the cylindrical pellet with a diameter [of 3mm], and a die length of 3mm. After the obtained pellet measured the melt flow rate by 230 degrees C and 2160g load, it produced the 200mmx400mm plate with the injection molding machine of 75t of clamping pressure, measured gloss and a surface-electrical-resistance value, and checked the existence of the flow mark visually further. Moreover, the eye ZODDO test piece with a thickness of 5mm was fabricated to coincidence, and eye ZODDO impact strength was measured.

[0031] The gay PP of the [example 4] example 3 was changed into Random PP, and also the same conduct as an example 1 was performed.

[0032] The gay PP of the [example 4 of comparison] example 3 was changed into Block PP, and also the same conduct as an example 1 was performed.

[0033] The pitch (Gay PP and Metallocene LLDPE) of the [example 5 of comparison] example 3 was changed into TPO. Others performed the same conduct as an example 1.

[0034] The metallocene LLDPE of the [example 6 of comparison] example 3 was changed into LLDPE in which the polymerization was carried out by the Ziegler catalyst. Others performed the same conduct as an example 1.

[0035]

[Table 1]

表1 材料の説明

略 号	内 容	M. F. R.
ホモPP	グラントポリプロ J 108M、ホモポリマー グラントポリマー社製	40
ランダムPP	グラントポリプロ J 226EA、ランダムコ ポリマー グラントポリマー社製	20
ブロックPP	グラントポリプロ J 708、ブロックコポリ マー グラントポリマー社製	40
TPO	ミラストマーM3800N、EPDM含有PP エラストマー 三井化学社製	15
メタロセンLL	カーネル KJ740、メタロセンLLDPE 日本ポリケム社製	80 60 (190℃)
チーグラールLL	ウルトゼックス3021F、チーグラールLLD PE 三井化学社製	3.3 2.1 (190℃)
カーボン	導電性カーボンブラック パルカン9A-32 キャボット社製	=====
金属酸化物	SbドーブSnO2 SN-100P 石原産業社製	=====
酸化防止剤	イルガノックス1076、フェノール系酸化防 止剤 チバ・スペシャルティケミカル社製	=====

* M. F. R.: 230℃、2160g荷重にて測定した。単位: g/10min
但しポリエチレン系樹脂は230℃、2160g荷重と190℃、2160g荷重の
2点で測定した。

[0036]

[Table 2]

表2 実施例、比較例での各特性値測定結果 (導電材料: カーボンブラック)

	実-1	実-2	比-1	比-2	比-3
ホモPP	59.8				59.8
ランダムPP		59.8			
ブロックPP			59.8		
TPO				79.8	
メタロセンLL	20.0	20.0	20.0		
チーグラールLL					20.0
カーボン	20.0	20.0	20.0	20.0	20.0
酸化防止剤	0.2	0.2	0.2	0.2	0.2
表面抵抗値 [Ω]	2×10^4	8×10^4	1×10^4	4×10^4	5×10^4
Izod 衝撃値 [J/m]	30.0	31.5	34.0	35.0	31.0
M.F.R. [g/10min]	7.2	6.3	6.8	1.1	2.3
光沢 [-]	72.0	74.3	31.9	16.8	49.6
フローマーク [-]	なし	なし	なし	あり	あり

[0037]

[Table 3]

表3 実施例、比較例での各特性値測定結果 (導電材料: 金属酸化物)

	実-3	実-4	比-4	比-5	比-6
ホモPP	29.8				29.8
ランダムPP		29.8			
ブロックPP			29.8		
TPO				39.8	
メタロセンLL	10.0	10.0	10.0		
チーグラールLL					10.0
金属酸化物	60.0	60.0	60.0	60.0	60.0
酸化防止剤	0.2	0.2	0.2	0.2	0.2
表面抵抗値 [Ω]	6×10^4	5×10^4	1×10^4	8×10^4	5×10^4
Izod 衝撃値 [J/m]	19.0	22.5	23.0	27.0	21.0
M.F.R. [g/10min]	3.3	3.1	3.8	0.6	1.0
光沢 [-]	53.0	57.8	24.9	11.7	22.6
フローマーク [-]	なし	なし	若干	あり	あり

表2、3の評価試験方法

- * 表面抵抗値 : JIS K7194に規定された4探針法による測定器を使用し、表面抵抗値を測定した。
- * Izod 衝撃値 : JIS K7110 試験片厚さ=5mm、後ノッチ、ノッチタイプ=Aノッチ、温度23℃下にて測定した。
- * 光沢 : グロスメーター (入射角、反射角とも60°) にて、平板表面を測定した。
- * フローマーク : 平板表面を目視確認した。

In the conductive resin mold goods obtained in the examples 1-4, it turns out among the example of [explanation of Tables 2 and 3] 10 combination that gloss shows a good high appearance.

[0038] The gestalt and example of operation which were indicated this time are instantiation in all points, and are not restrictive. The range of this invention is shown by the above-mentioned not explanation but claim, and it is meant that all modification in a claim, equal semantics, and within the limits is included.

[0039]

[Effect of the Invention] Since the conductive resin constituent of this invention contains 10 - 80 % of the weight of polypropylene regins obtained by propylene homopolymerization or the random

copolymerization of a propylene and ethylene, 5 - 50 % of the weight of straight chain-like low density polyethylene by which the polymerization was carried out, using a metallocene catalyst as a modifier component, carbon black, graphite, a metal powder and its whisker, a metal fiber, a metallic oxide, and 5 - 70 % of the weight of at least one or more kinds of electrical conducting materials chosen from a carbon fiber, the fluidity of a conductive resin constituent and the resin at the time of the mold-goods manufacture becomes high, and it has a good moldability.

[0040] Moreover, since very cheap supply of the raw material of conductive resin mold goods can be performed when an electrical conducting material is carbon black in the conductive resin constituent of this invention, the effectiveness which was excellent in the cost side is brought about.

[0041] Moreover, since the straight chain-like low density polyethylene which is used in the conductive resin constituent of this invention and by which the polymerization was carried out using the metallocene catalyst is in the range of the melt flow rate measured value of 1-100g / 10min. in 230 degrees C and 2160g load, the fluidity of a conductive resin constituent and the resin at the time of the mold-goods manufacture becomes the optimal, and it has a good moldability.

[0042] Moreover, since 840 - 920 kg/m³ and a bending elastic modulus have the consistency of the straight chain-like low density polyethylene which is used in the conductive resin constituent of this invention and by which the polymerization was carried out using the metallocene catalyst in the range of 100-500MPa, it has the impact strength which was excellent in a conductive resin constituent and its mold goods.

[0043] It becomes possible to raise the fluidity of resin, and the surface appearance of mold goods, maintaining the shock resistance of mold goods by using Metallocene LLDPE as an impact amelioration agent into a conductive resin constituent, and considering as the homopolymer of polypropylene, or a random copolymer with ethylene as base resin as stated above.

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CLAIMS

[Claim(s)]

[Claim 1] 10 - 80 % of the weight of polypropylene resins obtained by propylene homopolymerization or the random copolymerization of a propylene and ethylene, 5 - 50 % of the weight of straight chain-like low density polyethylene by which the polymerization was carried out, using a metallocene catalyst as a modifier component, carbon black, graphite, a metal powder and its whisker, a metal fiber, a metallic oxide, the conductive resin constituent characterized by including 5 - 70 % of the weight of at least one or more kinds of electrical conducting materials chosen from a carbon fiber.

[Claim 2] The conductive resin constituent according to claim 1 characterized by an electrical conducting material being carbon black.

[Claim 3] The conductive resin constituent according to claim 1 or 2 with which the straight chain-like low density polyethylene by which the polymerization was carried out using the metallocene catalyst is characterized by being in the range of the melt flow rate measured value of 1-100g / 10min. in 230 degrees C and 2160g load.

[Claim 4] claims 1-3 characterized by the straight chain-like low density polyethylene by which the polymerization was carried out using the metallocene catalyst being in the range of a consistency 840 - 920 kg/m³, and bending elastic-modulus 100-500MPa -- either -- the conductive resin constituent of a publication.

[Claim 5] a claim -- the mold goods obtained using the conductive resin constituent of a publication one to 4 either.

[Translation done.]